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Comparison between continued-fraction and computer-simulation methods applied to a classical Heisenberg chain

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A detailed comparison between the simulation data of Heller and Blume, and various continued-fraction expansions, is presented. It is shown that the three-pole expansion is qualitatively wrong while four- and six-pole expansions are in good agreement with the simulation data. A possibility of using continued-fraction representations to interpret simulation results is discussed. The convergence of the various approximations in the frequency domain is investigated.

I. INTRODUCTION

The properties of a classical Heisenberg chain have been studied quite extensively. This model has the interesting feature that the statics can be solved exactly[1] while the dynamics are not trivial.

As there is no long-range order for $T > 0$ in such a magnet,[2] it is an ideal system to study the spin dynamics in the paramagnetic region over a wide temperature range.

Motivated by the experimental work on TMMC[3] (tetramethylammonium-trichloromanganate, a very good antiferromagnetic $S = \frac{5}{2}$ Heisenberg chain) most theoretical work deals with the problem of obtaining an expression for the dynamical-structure factor for inelastic neutron scattering. Thereby, one assumes that, because of the high-spin value, it is a good approximation to replace the quantum-mechanical model by a classical one. The comparison between theory and experiment is primarily qualitative.

The classical Heisenberg chain is an ideal model for a computer-simulation study[3,4] and because simulation and theoretical methods deal with exactly the same model, these numerical experiments can be used to test the theoretical predictions. On the other hand, a direct comparison between simulations and experiments is not straightforward because experiments deal with the low-frequency regime whereas simulations are restricted to relatively short times (high frequencies).

Essentially, the theoretical methods to treat the dynamics can be divided in two classes. One approach is based on the cumulant expansion.[5,6] In this way, the time-dependent spin-spin correlation function is calculated directly and a comparison with computer-simulation data shows that, except for very short times, there is no quantitative agreement.[5,7]

Most authors use Mori's formalism[8] as a starting point for a description of the dynamics. In this way, an exact continued-fraction representation for the Laplace-transformed correlation function is derived. The frequency moments, appearing in this expansion, can be calculated rigorously.[9,10] The remaining problem, for which a variety of approximative methods can be found in the literature,[9–12] is the evaluation of the memory function.

As a final expression of such a calculation is frequency dependent, it is easy to make a qualitative comparison with experimental results. However, for a confrontation with numerical experiments one first has to perform an inverse Laplace transformation.

Recent neutron scattering experiments[13] demonstrated that the approximations proposed in Ref. 9 and 10 are qualitatively wrong. Indeed, these theories predict a central resonance for certain temperatures and wave vectors which is in complete contradiction with experimental observations.[13]

A comparison between the theory of Ref. 10 and the simulation data can be found in Ref. 7. Any continued-fraction representation exactly fulfills a number of frequency sum rules and this implies that the short-time behavior is correctly given by the inverse Laplace transform for the continued fraction. Therefore, it can be expected that there is perfect agreement between simulation and theory for very short times. For longer times there is no quantitative agreement.[7,9]

In Ref. 11, De Raedt and the present author showed how to approximate the memory function as consistently as possible and demonstrated that the qualitative agreement between experiment and theory increases drastically if the continued fraction is terminated at a later stage.

While the approximations, proposed in Ref. 9–11, are far from rigorous, Reiter and Sjölander obtained an exact perturbative expression for the memory function.[12] Their results, which are asymptotically correct for $T \rightarrow 0$, are in good agreement with the computer-simulation data for short times.

The purpose of this paper is twofold. (i) We want to compare our theory with the computer experiments because, recently, considerable progress has been made in extending both the accuracy and the time interval of these simulations.[14] Indeed, a comparison between our theory and the previous simulation data would hardly allow us to draw a conclusion. The time intervals for which data are available are short and since the theory exactly fulfills four sum rules, the short-time behavior would certainly agree within the rather large statistical errors. Therefore, we only present a detailed comparison between the data of Heller and Blume[14] and our theoretical results. (ii) The theory will be extended to include up to the tenth frequency moment[15] and the convergence of the results as a function of the number of moments will be investigated.

The plan of the paper is as follows: In Sec. II, we briefly recall the basic theory. In Sec. III, we compare the time-dependent spin-spin correlation functions, obtained from the continued-fraction representation, with the simulation data. In Sec. IV, the convergence of successive continued-fraction approximations is discussed. The conclusions of the paper are summarized in Sec. V.

II. BASIC THEORY

The Hamiltonian of a one-dimensional Heisenberg magnet is given by

$$H = -J \sum_q \vec{S}_q \cdot \vec{S}_{-q} \cos q \quad . \tag{2.1}$$

In the following, the spin operators will be replaced by classical vectors because then all static quantities can be calculated exactly.[1,9,10,15]

The normalized Laplace-transformed relaxation function is defined by

$$\Phi(z,q) = -i \int_0^\infty e^{izt} \frac{\langle \vec{S}_{-q}(t) \cdot \vec{S}_q(0) \rangle}{\langle \vec{S}_{-q}(0) \cdot \vec{S}_q(0) \rangle} dt,$$
$$z = \omega + i\epsilon, \quad \epsilon > 0 \quad . \tag{2.2}$$

This function can be written in an exact continued-fraction representation[8]

$$\Phi(z,q) = \cfrac{1}{z - \cfrac{\Delta_1(q)}{z - \cfrac{\ddots \quad \Delta_{N-1}(q)}{z - \Delta_N(q)\Phi^{(N)}(z,q)}}} \quad , \tag{2.3}$$

whereby $\Delta_1(q), \ldots, \Delta_N(q)$ are related to the frequency moments of the imaginary part of $\Phi(z,q)$. It is straightforward but very tedious to express the frequency moments in terms of static correlation functions and consequently only a limited number of Δ's have been calculated exactly. Therefore, one has to make some approximations for the higher-order relaxation functions $\Phi^{(N)}(z,q)$.

Here, the method proposed in Ref. 11 will be used, because for any value of N, the calculation of $\Phi^N(z,q)$ is simple and systematic. Furthermore, no adjustable parameters have to be introduced. In the first approximation, the final result of such a calculation reads[11]

$$\Phi^{(N)}(z,q) = [z + i\tau_N^{-1}(q)]^{-1} \quad . \tag{2.4}$$

The relaxation time $\tau_N(q)$ can be expressed as a function of the frequency moments and explicit expressions for $2 < N < 6$ are given in Appendix A.

Regardless of the approximation made for $\Phi^{(N)}(z,q)$, it is easy to make a qualitative comparison with experimental results.[9–11] In the classical limit, the dynamic-form factor for inelastic-neutron scattering is given by

$$S_{\rm cl}(\omega,q) = -(1/\beta)\Phi''(\omega,q) \quad ,$$
$$\Phi''(\omega,q) = \lim_{\epsilon \to 0} \mathrm{Im}\Phi''(z,q) \quad . \tag{2.5}$$

Assuming that most quantum mechanical effects are taken into account by introducing the Bose factor, the real dynamical structure factor is approximated by[3]

$$S(\omega,q) = \{\beta\omega/[1-\exp(-\beta\omega)]\} S_{\rm cl}(\omega,q) \quad . \tag{2.6}$$

It should be noted that such a comparison is qualitative only because $S(q,\omega)$ has to be convoluted with the experimental resolution function before a detailed comparison is possible. A lot of such qualitative comparisons can be found in the literature,[3,9–11] and we refer the interested reader to the original papers.

Starting from the continued-fraction representation Eq. (2.3) and the approximate expression for $\Phi^{(N)}(z,q)$ given by Eq. (2.4), it is quite simple to calculate the time-dependent correlation function $\langle \vec{S}_{-q}(t) \cdot \vec{S}_q(0) \rangle$. By means of some straightforward algebraic manipulations, the approximated $\Phi(z,q)$ can be written as a ratio of two polynomials

$$\Phi(z,q) = \sum_{n=0}^{N} \alpha_n(q) z^n / \sum_{n=0}^{N+1} \beta_n(q) z^n \quad . \tag{2.7}$$

For convenience, Eq. (2.7) is rewritten

$$i\Phi(is,q) = \sum_{n=0}^{N} a_n(q) s^n / \sum_{n=0}^{N+1} b_n(q) s^n \quad . \tag{2.8}$$

Of course, the coefficients a_n and b_n are completely determined by the Δ's and τ_N^{-1}. Explicit expressions can be found in Appendix A. The inverse Laplace transform of Eq. (2.8) is given by

$$\frac{\langle \vec{S}_{-q}(t)\cdot\vec{S}_q(0)\rangle}{\langle \vec{S}_{-q}(0)\cdot\vec{S}_q(0)\rangle}=\sum_{j=1}^{N+1} c_j(q)e^{-X_j(q)t}\ , \tag{2.9}$$

where

$$c_j(q)=\sum_{n=0}^{N} a_n(q)X_j^n(q)\Big/\sum_{n=0}^{N}(n+1)b_n(q)X_j^n(q)\ ,$$

and $X_j(q), j=1,\ldots,N+1$ denote the roots of the equation

$$\sum_{n=0}^{N+1} b_n(q)s^n=0\ . \tag{2.10}$$

In practice, $X_j(q)$ and $c_j(q)$ are most easily calculated by means of a simple but accurate computer program. Once X_j and c_j are known, the Fourier-transformed relaxation function

$$\Phi''(\omega,q)=\int_{-\infty}^{+\infty} e^{i\omega t}\frac{\langle \vec{S}_{-q}(t)\cdot\vec{S}_q(0)\rangle}{\vec{S}_{-q}(0)\cdot\vec{S}_q(0)\rangle}dt \tag{2.11}$$

is given by

$$\Phi''(\omega,q)=\sum_{j=1}^{N+1}\frac{c_j(q)X_j(q)}{\omega^2+X_j^2(q)}\ . \tag{2.12}$$

This expression suggests that $\Phi''(\omega,q)$ can be approximated by some superposition of Lorentzians. Of course, this is not so, because for a given N, $\Phi''(\omega,q)$ exactly fulfills the frequency sum rules

$$\langle\omega^{2n}\rangle_q=-\frac{1}{\pi}\int_{-\infty}^{+\infty} d\omega\,\omega^{2n}\Phi''(\omega,q),\quad n=0,\ldots,N\ . \tag{2.13}$$

Substituting Eq. (2.12) into Eq. (2.13) would certainly yield divergent results if there are no special relations between the X_j and c_j.

III. COMPARISON WITH COMPUTER SIMULATION

Starting from the basic formulas (2.9) and (2.10) and the expressions given in Appendix A, it is clear which steps have to be made in order to obtain the time-dependent spin-spin correlation function. For a given temperature and wave vector, we calculate the moments $\langle\omega^{2n}\rangle_q$, $1\leq n\leq N$. Then the coefficients $a_n(q)$ and $b_n(q)$ are calculated and the algebraic Eq. (2.10) is solved numerically. Once the roots $X_j(q)$ have been found, the coefficients $c_j(q)$ are known and consequently the time-dependent correlation function can be plotted as a function of time. In Fig. 1–5 we compare our results for $2<N<6$ for a fixed temperature and various wave vectors with the simulation data of Heller and Blume.[14] Here, we only present results for the antiferromagnet because the general conclusions drawn from this case also hold for the ferromagnet.

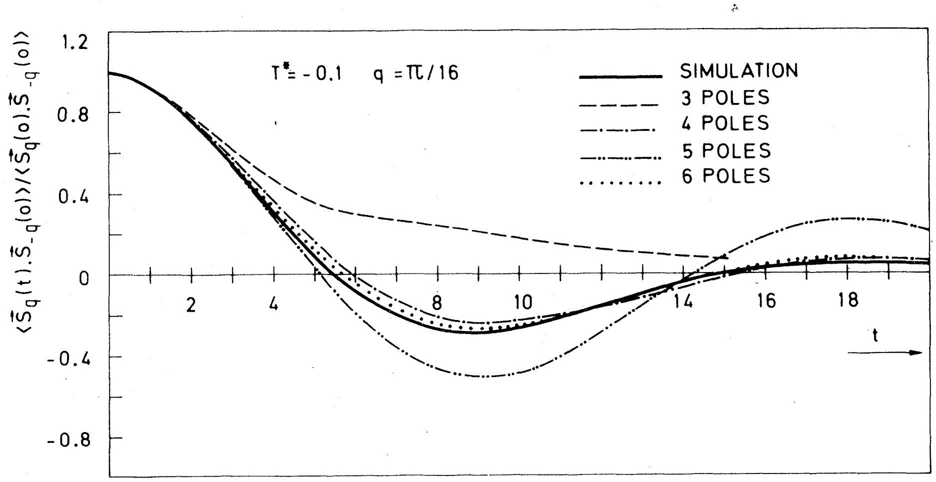

FIG. 1. Normalized time-dependent correlation function obtained from continued-fraction expansions compared to the simulation data of Heller and Blume. In all figures, $T^*=T/JS(S+1)$ where T denotes the temperature. Time is measured in units of $|J|$.

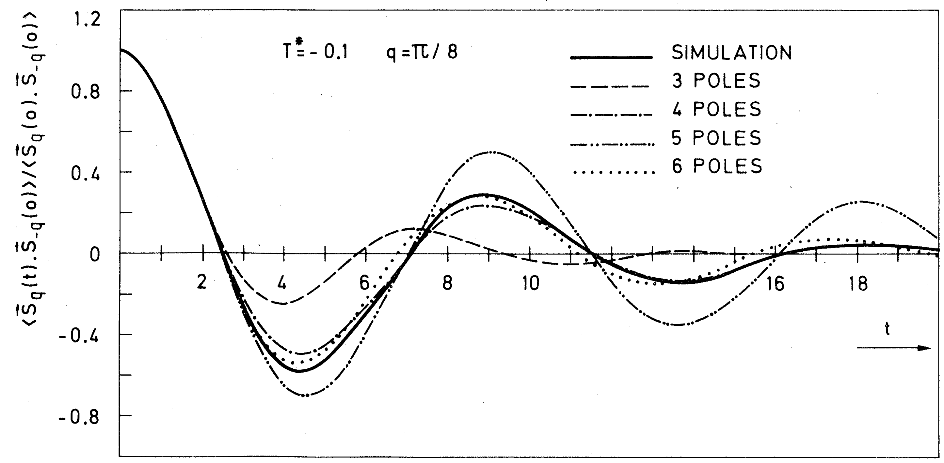

FIG. 2. Normalized correlation function for $T^* = -0.1$ and $q = \frac{1}{8}\pi$. Note the strange behavior of the three-pole expansion.

Looking at Fig. 1, it is obvious that the three pole expansion ($N=3$) is qualitatively wrong. This fact is closely related to the observation that the criterion to observe spin waves is $q >> \kappa^{1/2}$ (instead of $q >> \kappa$) for the three-pole expansion.[9,11] In order to investigate this point further, it is useful to make the low-temperature expansion of the moments and to compare the relaxation time τ_q^{-1} with the approximate spin-wave frequency $\omega_0(q) = (\langle \omega^2 \rangle_q)^{1/2}$. Indeed, most approximations for $\Phi^{(N)}(z,q)$ are based on the assumption that $\Phi^{(N)}(z,q)$ describes fast-fluctuating processes only.[8-11] A simple criterion for the validity of this assumption is thus $\omega_0(q)\tau_N(q) << 1$. Using the expressions given in Appendix B, we find

$$\omega_0^2(q)\tau_3^2(q) = 1 - \tfrac{1}{2}T(3+\cos q)/(1-\cos^2 q), \quad T \to 0 \ . \tag{3.1}$$

This implies

$$\tfrac{1}{2}T(3+\cos q)/(1-\cos^2 q) >> 0 \tag{3.2}$$

and therefore, this approximation is only valid if $q \to 0$ or $q \to \pi$. That this is no artifact of our method is most easily seen by taking the expression

$$\tau_{\rm LM}^{-2}(q) = \tfrac{1}{2}\pi\Delta_2(q) \tag{3.3}$$

proposed by Lovesey and Meserve.[10] Then we obtain

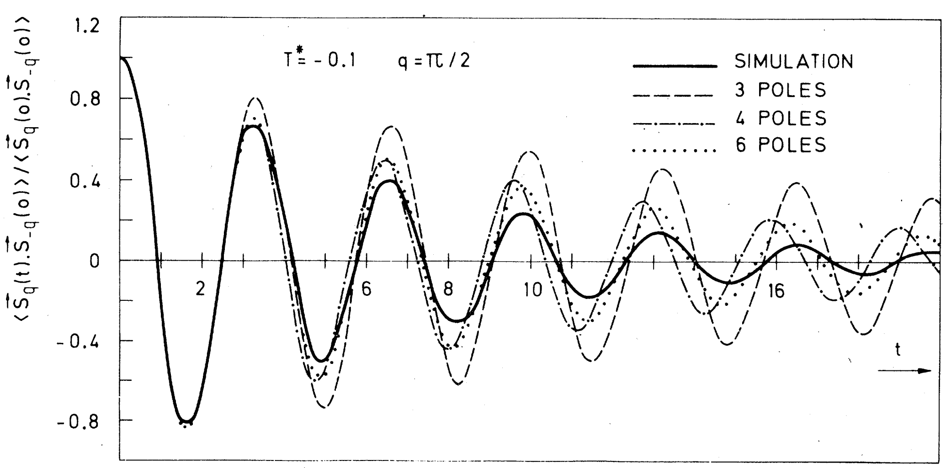

FIG. 3. Five-pole expansion is omitted because of clarity. In all cases, there is no quantitative agreement for $t > 3$. A similar behavior is found in the ferromagnetic case.

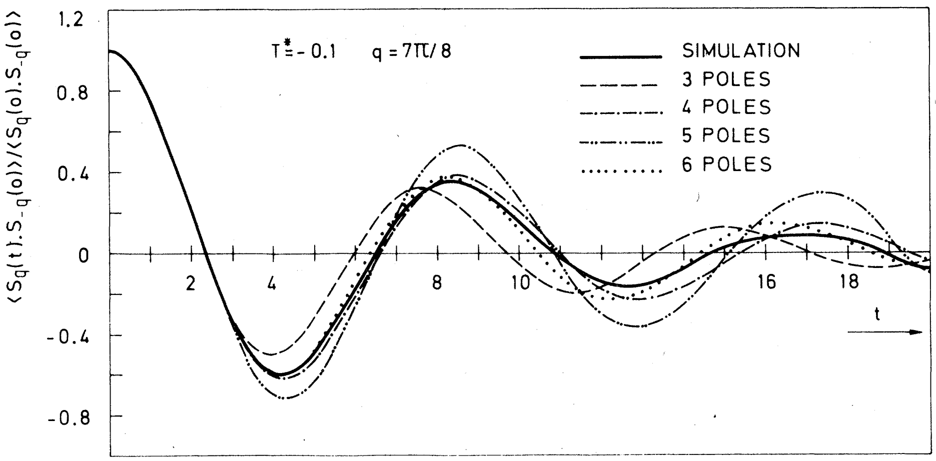

FIG. 4. Three-pole and five-pole expansions are qualitatively wrong.

the criterion

$$\cos q < -1+\tfrac{1}{4}\pi T \quad \text{or} \quad \cos q > 1-\tfrac{1}{2}\pi T \ , \tag{3.4}$$

which is essentially the same as in our case. In our opinion, the fundamental reason for the failure of this simple three-pole approximation can be seen if the first terms in the continued fraction are expanded to first order in T. Again, we use the expressions (B1–B4) and we obtain

$$\Phi(z,q)=\cfrac{1}{z-\cfrac{4(1-2T)(1-c^2)}{z-\cfrac{2T(3+c)}{z-\cfrac{2(1+c+2c^2)}{z-\cfrac{2(5+11c+25c^2+5c^3-10c^4-4c^5)/(3+c)(1+c+2c^2)}{z-\cdots}}}}} \tag{3.5}$$

where $c=\cos q$.

Clearly, the temperature dependence (for $T\rightarrow 0$) is entirely determined in the first two stages of the continued fraction. As the three-pole expansion is obtained by truncating the continued fraction at the second stage, one should not expect this representation to yield a good overall description of the dynamical properties of the Heisenberg chain.

From Fig. 1, 2, 4, and 5, we may conclude that there is an excellent agreement between four-pole,[16] six-pole, and simulation results. For $q=\frac{1}{2}\pi$ (Fig. 3), the inverse relaxation times $\tau_4^{-1}(q)$, $\tau_5^{-1}(q)$, and $\tau_6^{-1}(q)$ are of the same order of magnitude as the spin wave frequency and consequently, we should not expect our description to be quantitatively correct. It is surprising that there is a serious disagreement between the five-pole expansion results and other data. If the number of poles is odd, Eq. (2.10) has at

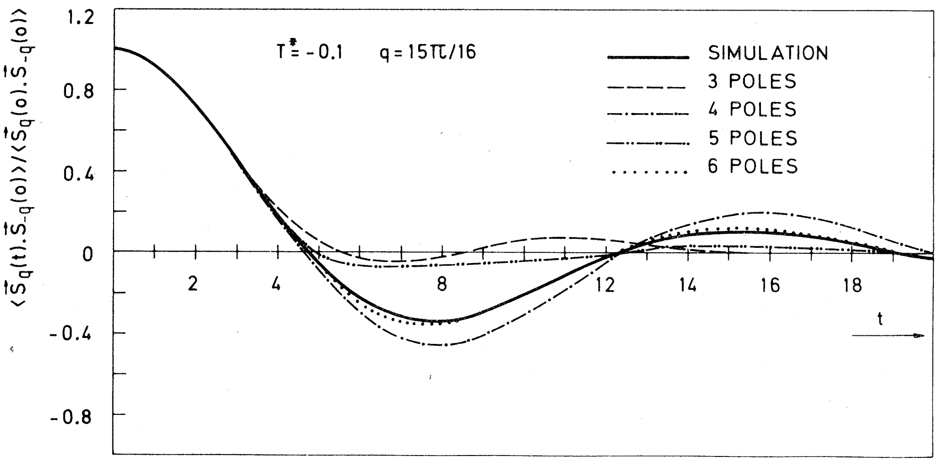

FIG. 5. Time-dependent correlation function for $q \approx \pi$. In general the six-pole expansion is better than the four-pole expansion, but the five-pole expansion is worse than the four-pole expansion.

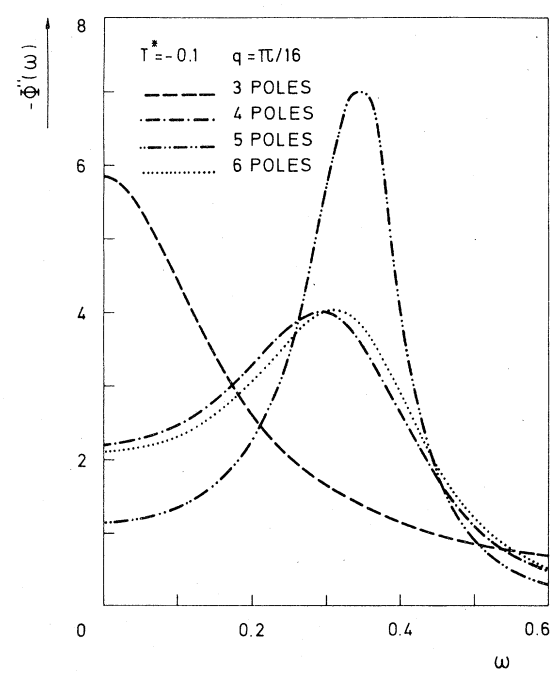

FIG. 6. Fourier-transformed correlation function for the case depicted in Fig. 1. Good agreement between four- and six-pole expansion in the time domain is clearly reflected.

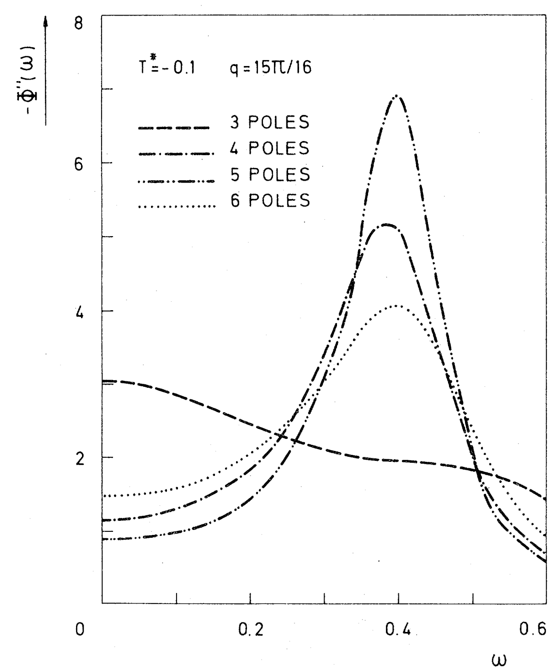

FIG. 7. Fourier transforms of the correlation functions shown in Fig. 5.

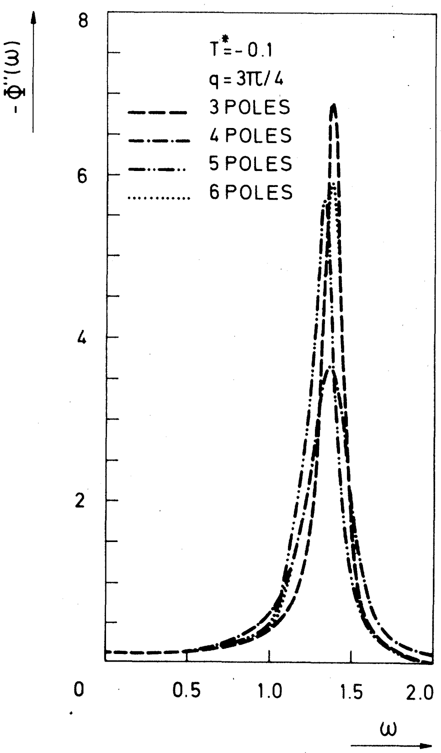

FIG. 8. In this case good qualitative agreement between three-, four-, five-, and six-pole expansion is found.

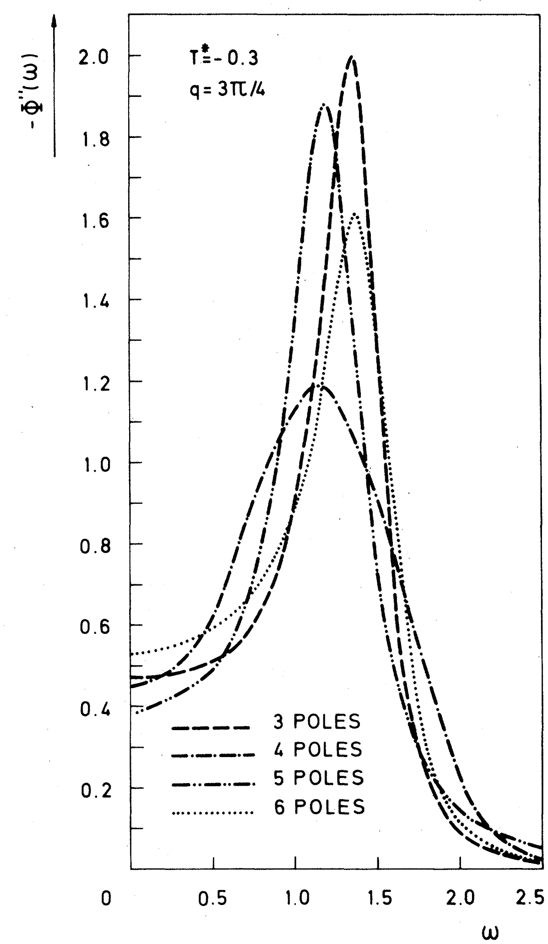

FIG. 9. By comparing these line shapes with the ones of Fig. 8 we may conclude that the agreement gets worse if the temperature is increased.

least one real solution. Such a pole corresponds to a pure-relaxation process. Therefore, it can be expected that it is difficult to approximate a function with dominant oscillatory behavior by means of a continued fraction with an odd number of poles.

We close Sec. III by discussing the possibility of using the continued-fraction representation to interpret simulation data in more complex situations. In general, the quantities $\Delta_1(q),\ldots,\Delta_N(q)$ can be calculated directly by averaging over the spin configurations obtained by a Monte Carlo process.[17] In this way, it is not necessary to express all Δ's in terms of static correlation functions. Of course, in the Heisenberg case, a number of Δ's can be calculated rigorously and therefore, these calculations can be omitted. Using $\tau_N(q)$ as an adjustable parameter, the time-dependent correlation function calculated from the continued-fraction representation can be fitted to the time-dependent correlation function obtained by integrating the equations of motion.[18]

Obviously, this procedure is much simpler than treating $\Delta_1(q),\ldots,\Delta_N(q)$ and $\tau_N(q)$ as fitting parameters.[7]. For practical applications, we feel that the four-pole expansion yields a good compromise between complexity and accuracy.

IV. FREQUENCY-DEPENDENT CORRELATION FUNCTIONS

In Sec. IV, we investigate the convergency of the continued-fraction representation as a function of the number of moments by means of a few representative examples.

In Figs. 6 and 7, the fact that the three-pole expansion is too simple is clearly illustrated by the presence of a central resonance. Of course, this could have been expected because Fig. 1 and 5 already demonstrated that the oscillations are strongly damped. The good agreement between the four-pole and six-pole expansions in the time domain results in a good qualitative agreement in the frequency domain. In general, the quantitative agreement is rather poor. From Figs. 8 and 9, we may conclude that the disagreement is worse for higher temperatures. This is easily understood because the approximations for $\Phi^{(N)}(z,q)$ are based on the distinction between slow and fast processes. For high temperatures only conserved quantities are slow and therefore, we should not expect these approximations to work well for $q \gg 0$.

In Fig. 10, we show an extreme case. Here, the six-pole expansion is definitely wrong. To analyze this case in more detail, it is useful to make a plot of the poles in the complex plane and to draw circles with radii τ_N^{-1}. Essentially the approximations for $\Phi^{(N)}(z,q)$ are "long-time" approximations[8] and one expects the approximation to yield a good description if all poles lie within the corresponding circle. Looking at Fig. 11, it is obvious that this is not the case for the six-pole expansion. Thus, we conclude that our approximation for τ_6^{-1} is no longer valid.

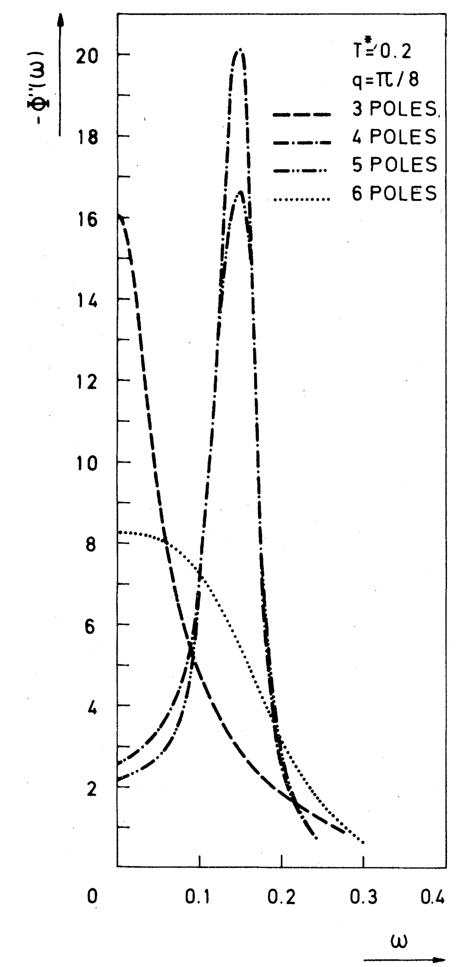

FIG. 10. Special case showing that the six-pole expansion is not always in good agreement with the four-pole representation.

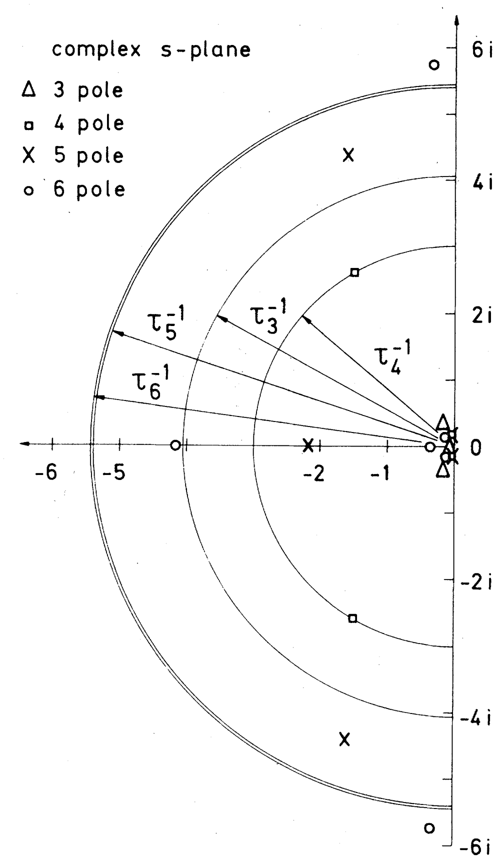

FIG. 11. Plot of the poles corresponding to the line shapes shown in Fig. 10.

In general, we conclude that the four- and six-pole expansion are in good qualitative agreement if there is a clear distinction between slow and fast processes. This is the case at low temperatures. The fact that our theory[11] compares well with the experimental results for TMMC can be readily understood if one notes that $T^* = T/JS(S+1) = -0.1$ corresponds to a temperature of about 11.4 K.

V. CONCLUSION

We have presented a detailed comparison between computer-simulation data and theoretical results, based on a continued-fraction representation of the time-dependent spin-correlation functions.

In general, the agreement between the four-pole and six-pole expansion results and the numerical data is good. Then we investigated the convergency of the continued-fraction expansions as a function of the number of poles and we found that the quantitative agreement gets worse if the temperature increases. Qualitatively, the three-pole expansion turns out to be wrong.

Finally, we have shown that a careful analysis of the poles is necessary to interpret the results of a continued-fraction expansion correctly.

ACKNOWLEDGMENTS

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APPENDIX A

It is straightforward to express Δ_j in terms of moments. The calculation of the relaxation times $\tau_N(q)$ is extensively discussed in Ref. 11 and therefore, only the final results are given.

Denoting the $2n$th frequency moment by $\langle\omega^{2n}\rangle_q$, we have

$$\Delta_1(q)=\langle\omega^2\rangle_q,\quad \Delta_2(q)=\langle\omega^4\rangle_q/\langle\omega^2\rangle_q-\langle\omega^2\rangle_q,\quad \tau_2^{-2}(q)=\langle\omega^4\rangle_q/\langle\omega^2\rangle_q \;,$$

$$\Delta_3(q)=(\langle\omega^6\rangle_q/\langle\omega^2\rangle_q-\langle\omega^4\rangle_q^2/\langle\omega^2\rangle_q^2)/\Delta_2(q) \;,$$

$$\tau_3^{-2}(q)=(\langle\omega^6\rangle_q/\langle\omega^2\rangle_q-2\langle\omega^4\rangle_q+\langle\omega^2\rangle_q^2)/\Delta_2(q) \;,$$

$$\tau_4^{-2}(q)=(\langle\omega^8\rangle_q/\langle\omega^2\rangle_q-2\langle\omega^6\rangle_q\langle\omega^4\rangle_q/\langle\omega^2\rangle_q^2+\langle\omega^4\rangle_q^3/\langle\omega^2\rangle_q^3)/\Delta_2(q)\Delta_3(q) \;,$$

$$\Delta_4(q)=\tau_4^{-2}(q)-\Delta_3(q) \;,$$

$$\tau_5^{-2}(q)=\{\langle\omega^{10}\rangle_q/\langle\omega^2\rangle_q-\langle\omega^8\rangle_q(\tau_3^{-2}+\langle\omega^2\rangle_q)/\langle\omega^2\rangle_q+\langle\omega^6\rangle_q[\tau_3^{-2}-\Delta_2(q)]\}/$$
$$[\langle\omega^8\rangle_q/\langle\omega^2\rangle_q-2\langle\omega^6\rangle_q\langle\omega^4\rangle_q/\langle\omega^2\rangle_q^2+\langle\omega^4\rangle_q^3/\langle\omega^2\rangle_q^3-\Delta_2(q)\Delta_3^2(q)]-\tau_3^{-2}(q)-\langle\omega_q^2\rangle \;,$$

$$\Delta_5(q)=\tau_5^{-2}(q)-\Delta_4(q) \;.$$

For any N, it is an easy exercise to express the coefficients $a_n(q)$ and $b_n(q)$ in terms of Δ's and τ's.

We obtain for $N=2$,

$$a_0(q)=\Delta_2(q) \;,$$
$$a_1(q)=\tau_2^{-1}(q) \;,$$
$$a_2(q)=1 \;,$$
$$b_0(q)=\tau_2^{-1}(q)\Delta_1(q) \;,$$
$$b_1(q)=\Delta_2(q)+\Delta_1(q) \;,$$
$$b_2(q)=\tau_2^{-1}(q) \;,$$
$$b_3(q)=1 \;,$$

for $N=3$,

$$a_0(q)=\tau_3^{-1}(q)\Delta_2(q) \;,$$
$$a_1(q)=\Delta_3(q)+\Delta_2(q) \;,$$
$$a_2(q)=\tau_3^{-1}(q) \;,$$
$$a_3(q)=1 \;,$$
$$b_0(q)=\Delta_1(q)\Delta_3(q) \;,$$
$$b_1(q)=\tau_3^{-1}(q)[\Delta_2(q)+\Delta_1(q)] \;,$$
$$b_2(q)=\Delta_3(q)+\Delta_2(q)+\Delta_1(q) \;,$$
$$b_3(q)=\tau_3^{-1}(q) \;,\quad b_4(q)=1 \;,$$

for $N=4$,

$$a_0(q)=\Delta_2(q)\Delta_4(q) \;,$$
$$a_1(q)=\tau_4^{-1}(q)[\Delta_3(q)+\Delta_2(q)] \;,$$
$$a_2(q)=\Delta_4(q)+\Delta_3(q)+\Delta_2(q) \;,$$
$$a_3(q)=\tau_4^{-1}(q) \;,$$
$$a_4(q)=1 \;,$$
$$b_0(q)=\tau_4^{-1}(q)\Delta_1(q)\Delta_3(q) \;,$$
$$b_1(q)=\Delta_4(q)[\Delta_2(q)+\Delta_1(q)]+\Delta_3(q)\Delta_1(q) \;,$$
$$b_2(q)=\tau_4^{-1}(q)[\Delta_3(q)+\Delta_2(q)+\Delta_1(q)] \;,$$
$$b_3(q)=\Delta_4(q)+\Delta_3(q)+\Delta_2(q)+\Delta_1(q) \;,$$
$$b_4(q)=\tau_4^{-1}(q) \;,$$
$$b_5(q)=1 \;,$$

for $N=5$,

$$a_0(q)=\tau_5^{-1}(q)\Delta_4(q)\Delta_2(q) \ ,$$

$$a_1(q)=\Delta_5(q)[\Delta_3(q)+\Delta_2(q)]+\Delta_4(q)\Delta_2(q) \ ,$$

$$a_2(q)=\tau_5^{-1} \ [\Delta_4(q)+\Delta_3(q)+\Delta_2(q)] \ ,$$

$$a_3(q)=\Delta_5(q)+\Delta_4(q)+\Delta_3(q)+\Delta_2(q) \ ,$$

$$a_4(q)=\tau_5^{-1}(q) \ ,$$

$$a_5(q)=1 \ ,$$

$$b_0(q)=\Delta_5(q)\Delta_3(q)\Delta_1(q) \ ,$$

$$b_1(q)=\tau_5^{-1}(q) \times \{\Delta_4(q)[\Delta_2(q)+\Delta_1(q)]+\Delta_3(q)\Delta_1(q)\} \ ,$$

$$b_2(q)=\Delta_5(q)[\Delta_3(q)+\Delta_2(q)+\Delta_1(q)]+\Delta_4(q)[\Delta_2(q)+\Delta_1(q)]+\Delta_3(q)\Delta_1(q) \ ,$$

$$b_3(q)=\tau_5^{-1}(q)[\Delta_4(q)+\Delta_3(q)+\Delta_2(q)+\Delta_1(q)] \ ,$$

$$b_4(q)=\Delta_5(q)+\Delta_4(q)+\Delta_3(q)+\Delta_2(q)+\Delta_1(q) \ ,$$

$$b_5(q)=\tau_5^{-1}(q) \ ,$$

$$b_6(q)=1 \ .$$

APPENDIX B

Here we calculate the low-temperature expansions of the moments to first order in the temperature. For simplicity, we take $|J|=1$ and $S^2=1$. For $m>1$, the frequency moments can be written

$$\frac{\langle\omega^{2m}\rangle_q}{\langle\omega^2\rangle_q}=\frac{2}{y}\sum_{n,l,J}A_{lJ}^n \cos qnT^l y^J \ ,$$

where $y=\coth 1/T-T$. The coefficients A_{lJ}^n have been calculated by B. Nickel. To first order in T, we then obtain in the case of an antiferromagnet

$$\langle\omega^2\rangle_q=4(1-2T)(1-c^2) \ , \quad \text{(B1)}$$

$$\langle\omega^4\rangle_q/\langle\omega^2\rangle_q=4(1-c^2)-2T(1-c-4c^2) \ , \quad \text{(B2)}$$

$$\langle\omega^6\rangle_q/\langle\omega^2\rangle_q=16(1-c^2)^2+4T(-1+8c+27c^2-2c^3-16c^4) \ , \quad \text{(B3)}$$

$$\langle\omega^8\rangle_q/\langle\omega^2\rangle_q=64(1-c^2)^3+16T(4+23c+65c^2-5c^3-65c^4+2c^5+24c^6) \ , \quad \text{(B4)}$$

$$\langle\omega^{10}\rangle_q/\langle\omega^2\rangle_q=128(1-c^2)^4+16T(61+232c+602c^2+28c^3-651c^4+28c^5+476c^6-8c^7-128c^8) \ ,$$

where $c=\cos q$. For the ferromagnet, we find

$$\langle\omega^2\rangle_q=4(1-2T)(1-c)^2 \ ,$$

$$\langle\omega^4\rangle_q/\langle\omega^2\rangle_q=4(1-c)^2-2T(1-c)(1-4c) \ ,$$

$$\langle\omega^6\rangle_q/\langle\omega^2\rangle_q=16(1-c)^4-4T(1-c)^2(1-22c+16c^2) \ ,$$

$$\langle\omega^8\rangle_q/\langle\omega^2\rangle_q=64(1-c)^6+16T(1-c)^3\times(4+37c-58c^2+24c^3) \ ,$$

$$\langle\omega^{10}\rangle_q/\langle\omega^2\rangle_q=128(1-c)^8+16T(1-c)^4\times(61+188c-516c^2+440c^3-122c^4) \ .$$

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